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1 BACKGROUND – 3D electrochemistry for enhanced organic degradation

Conventional (2D) electrochemical oxidation (EO) of micropollutants in water is, despite capability of full mineralization, challenged on the efficiency. This is due to mass transfer limitations of the single pass conversions induced by the inherent low concentrations of micropollutants [1]. Conductive particles may, when placed within an electric field, be polarized generating reactive microelectrodes in bulk solution increasing the overall active electrode area, a concept known as 3D electrochemistry [2]. Carbon based materials (activated carbon (AC) and others) may be cost-efficient electrocatalysts for use in 3D electrochemical water treatment.

OBJECTIVES

- the aim of this study was to search for potential synergy of using AC and a new Fe_3C@N-GE–CNT material as electrocatalysts in 3D electrochemistry based on removal kinetics of single and combined processes.

2 THEORY, METHODS AND MATERIALS

Theoretical concept

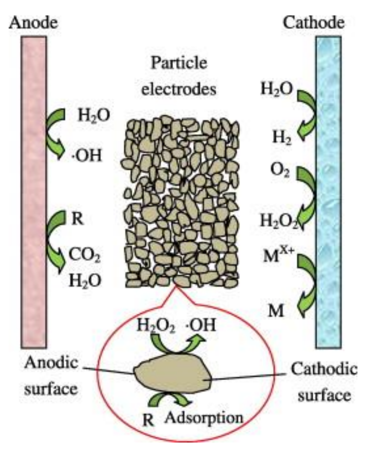


Figure 1: The reactions involved in 3D electrochemistry [1]

Batch setup

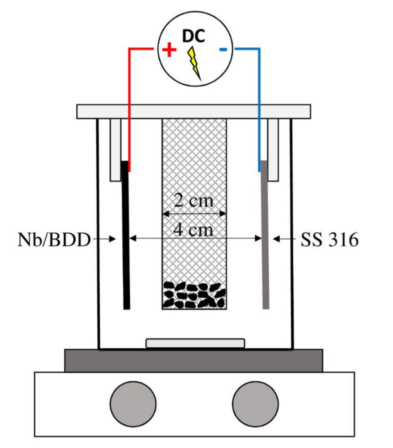
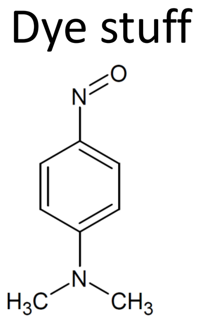


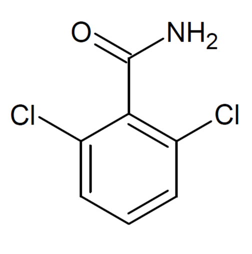
Figure 2: The batch cell in 2D (without) and 3D (with) particles [3]

Dye stuff

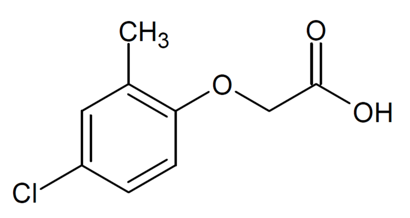


RNO

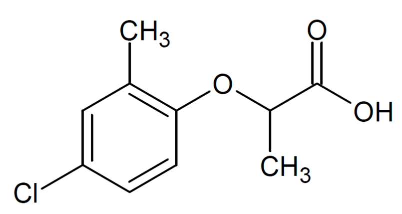
Pesticides and transformation product



BAM
log K_{ow} = 0.77
pK_a = 13-14



MCPA
log K_{ow} = 2.73
pK_a = 3.07



MCPP
log K_{ow} = 2.56
pK_a = 3.19

Synergy: $$S = \frac{k_{3D} - k_{2D} - k_{AC}}{k_{2D} + k_{AC}} \quad \text{Eq. 1}$$

Adsorption: $r = \frac{d[Org]}{dt} = -k_{Ad} \cdot [Org]$; $k_{Ad} = k \cdot [Carbon]_0$

2D Elec.: $r = \frac{d[Org]}{dt} = -k_{2D} \cdot [Org]$; $k_{2D} = k \cdot [OH^\bullet]_0$

3D Elec.: $r = \frac{d[Org]}{dt} = -k_{3D} \cdot [Org]$; $k_{3D} = k \cdot [OH^\bullet]_0 \cdot [Carbon]_0$

3 RESULTS – Process optimization (AC on Dye)

AC adsorption

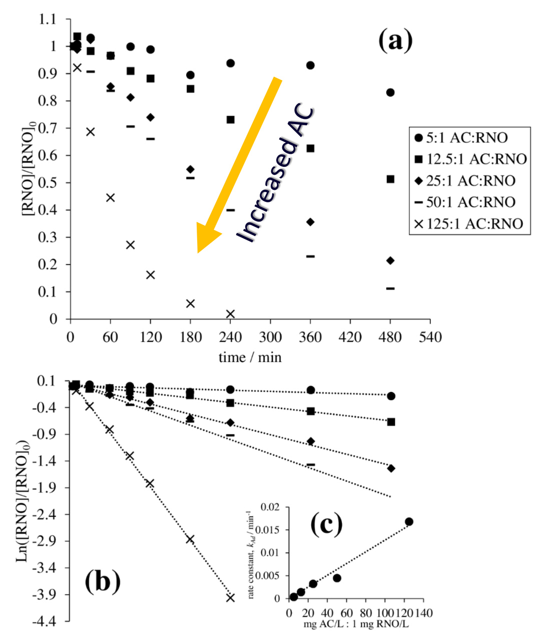


Figure 3: (a) RNO removal vs. w/w AC:RNO ratios. (b) First order plot, (c) k_{app} vs. constants vs. amount of AC added.

2D electrochemistry

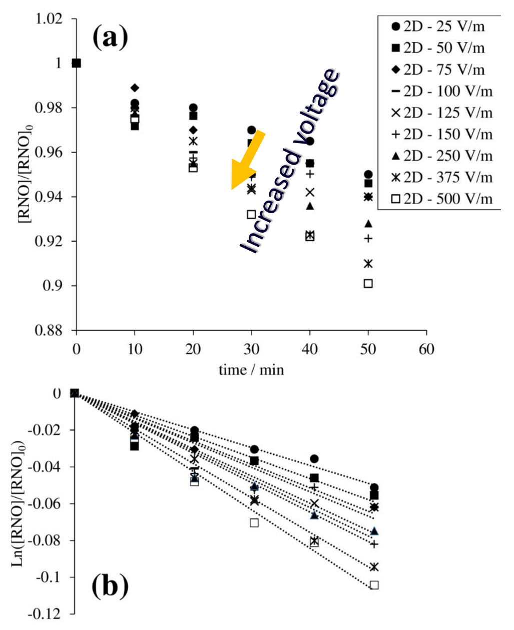


Figure 4: (a) 2D potentiostatic RNO removal (1 – 20 V) (b) First order plot.

3D electrochemistry

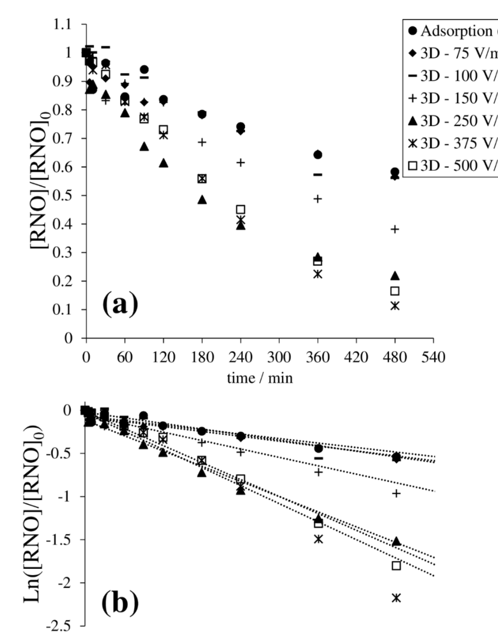
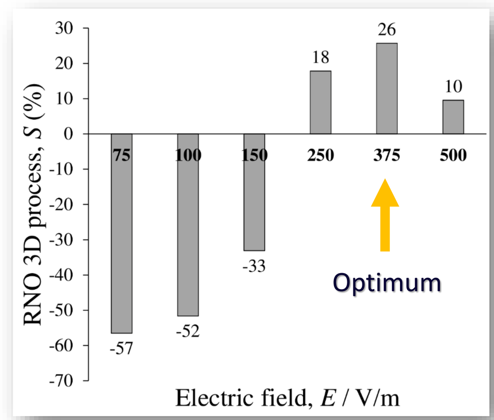


Figure 5: (a) 3D potentiostatic RNO removal (0 – 500 V/m) and AC:RNO 5:1 (b) First order plot.

Figure 6: Synergy vs. Electric field strength

Data presented is published by Pedersen et al. [3]



>250 V/m needed for GAC polarization

Optimum obtained at 375 V/m

4 RESULTS – Pesticide treatment using AC particles

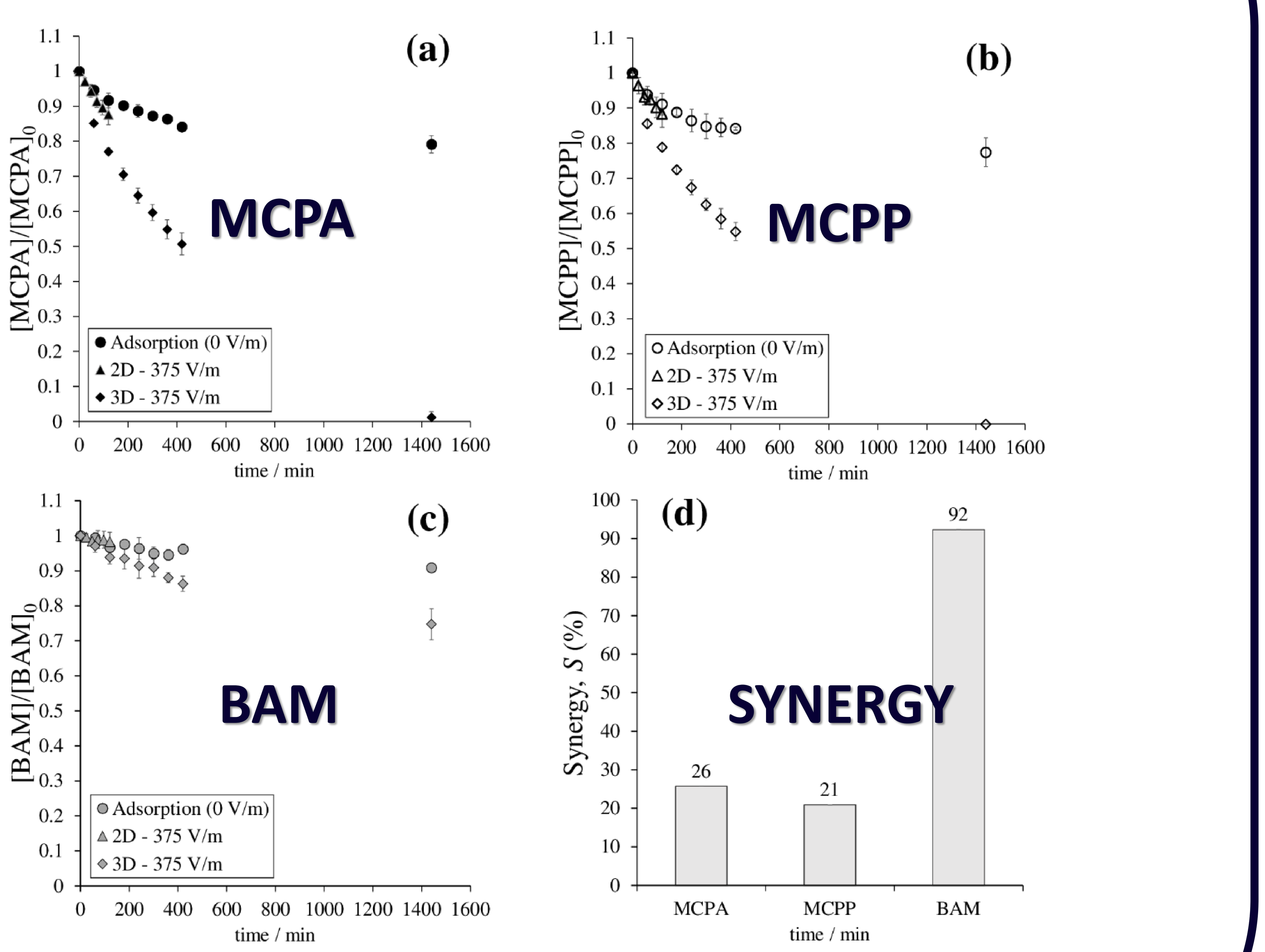


Figure 7: Degradation data of the MCPA, MCPP and BAM synergy study. (a) MCPA removal, (b) MCPP removal (c) BAM removal (d) Synergy of the processes. Electric field strength, E = 375 V/m, 100 mg AC/L, $[MCPA]_0$ = 50 mg/L, $[MCPP]_0$ = 50 mg/L, $[BAM]_0$ = 50 mg/L. Data presented is published by Pedersen et al. [3]

5 RESULTS – 3D electrochemistry with NCPE

Synthesis of Fe_3C@N-GE-CNTs (NCPE)

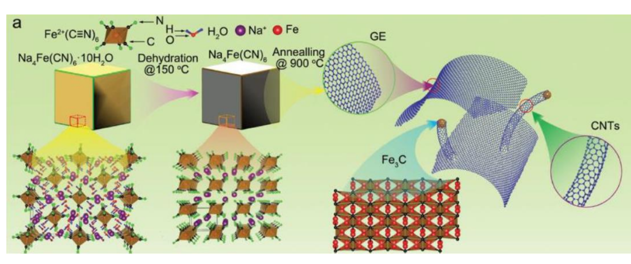
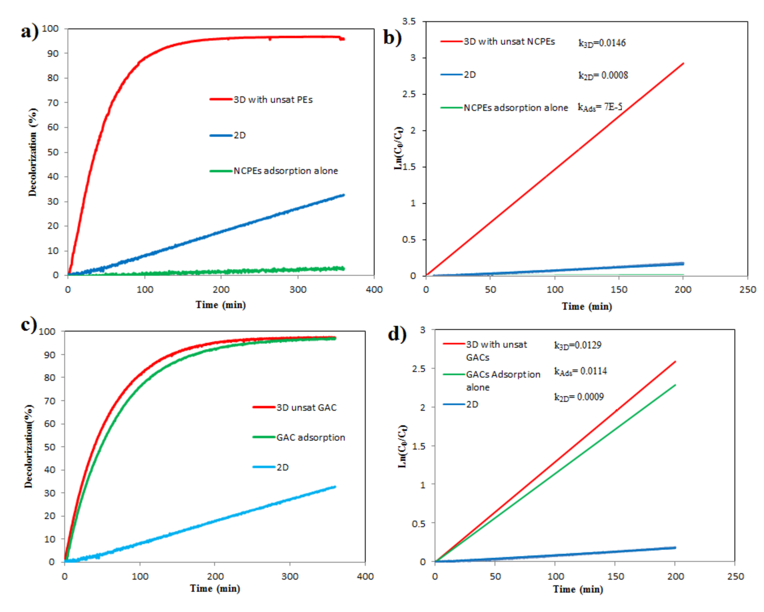


Figure 8: Illustration of the synthesis process for the Fe_3C@N-GE–CNTs hybrid particles from commercial $Na_4Fe(CN)_6$. The protocol is published by Su et al. [4]

NCPE vs. AC as electrocatalysts (on Dye)



NCPE

AC

Figure 9: 3D electrochemical RNO bleaching tests on NCPE (a and b) and activated carbon (c and d).

Activated carbon: Removal dominated by adsorption (low S)

NCPE: Removal dominated by reactive materials (high S)

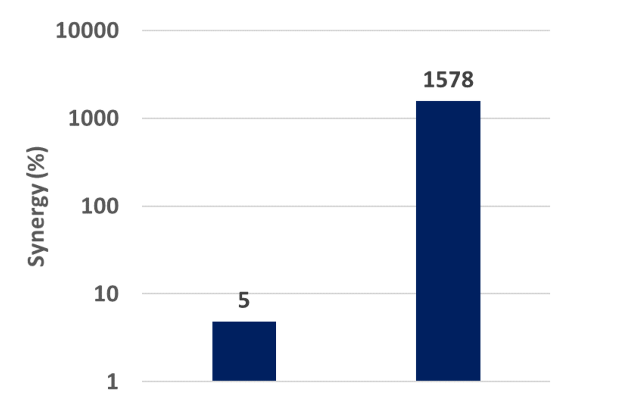


Figure 10: Calculated synergies of the bleaching according to equation 1.

CONCLUSIONS

- 3D electrochemistry provides synergism between adsorption and electrochemical degradation of micropollutants
- The dominant removal process of the system depends on the properties of the carbon based electrocatalyst (AC: adsorption ; NCPE: degradation)

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